A Phenomenological Study of Contamination Enhanced Laser-Induced Damage in Sealed Lasers

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***Abstract*- This research demonstrates that adding trace levels of contaminants to an otherwise evacuated system, leads to rapid onset of damage to optical elements in the presence of 1064 nm laser radiation.**

Specifically, 1064 nm radiation from a pulsed laser having approximately 800 mJ/cm^2 average power (<1.5 J/cm^2 peak power) illuminated fused silica windows used to seal a vacuum chamber, with a pressure <1.0×10^{-3} torr. In the absence of any contamination the windows were demonstrated to show no signs of damage up to 700,000 laser pulses. When gas phase toluene was introduced into the system at varying concentrations (<1.0×10^{-3} - 3.1×10^{-1} torr), the onset of damage was seen to be a function of the toluene concentration, and damage was seen to occur as rapidly as 30,000 laser pulses.

This phenomenon was also observed when the windows had a commercially applied coating of MgF_2 applied to the surface in the vacuum system Similar experiments using acetone as the contaminant led to no observed damage for either optic, even at high concentrations.

A discussion of possible mechanisms leading to damage is also included.

I. Introduction

The use of sealed, space-based lasers (SBL) by NASA for Earth Sciences studies includes LIDAR missions such as ICESAT[1], and the potential exists for various spectroscopic studies. In addition, planetary missions using LIDAR[2] have also incorporated sealed lasers, and other studies such as Laser Induced Breakdown Spectroscopy (LIBS) are proposed [3].

While the challenges of developing and constructing such instruments are immense, one unexpected and quite non-intuitive challenge to using sealed lasers is particularly notable, that is laser-induced damage of optics due to contamination[4]. This phenomenon was first reported in the literature with tests being conducted on sealed systems filled with ultra-pure nitrogen. With some contaminants, damage was seen to occur as quickly as 13,000 laser pulses for contamination levels of 175 ppm. Contaminants that led to rapid onset of damage included various aromatic hydrocarbons and silicones.

It is suggested that this effect is non-intuitive because these contaminants at the same or higher concentrations have had no reported effects on laboratory lasers through the years. One important conclusion from the reported work was that this effect was demonstrated only in the absence of (molecular) oxygen.

Here we present research where this work is extended to a vacuum environment, more representative of spacecraft environment Fused silica windows, both bare and with a MgF_2 coating, were tested for a changed 1064 nm laser-induced damage threshold due to the addition of contaminants.

II. Experimental

The test facility consists of a vacuum chamber, a contaminant reservoir, and the laser. A block diagram of the set-up is presented in Fig. 1. A six-way cross served as the vacuum chamber, and allowed for ready attachment of the other experimental components. The chamber was pumped from the bottom line of the cross using an oil free pump (The Pump Works, HV8). The contaminants were introduced individually from a side flange, perpendicular to the laser beam. During the experiments they are stored in a vacuum sealed stainless-steel reservoir, and trace amounts are bled into the system using a needle valve. The pressure of the system and the amount of contaminant added were monitored with a thermocouple gauge.

A window served as both the test optic and allowed the laser into of the system. A second window allowed the laser to exit the chamber, to prevent additional effects from interactions with the chamber walls. Typically, fused silica windows were used, (GM Associates, 7500-20, maximum scratch dig 80/20) with the inlet window serving as the test sample. During the course of this work, a few MgF_2

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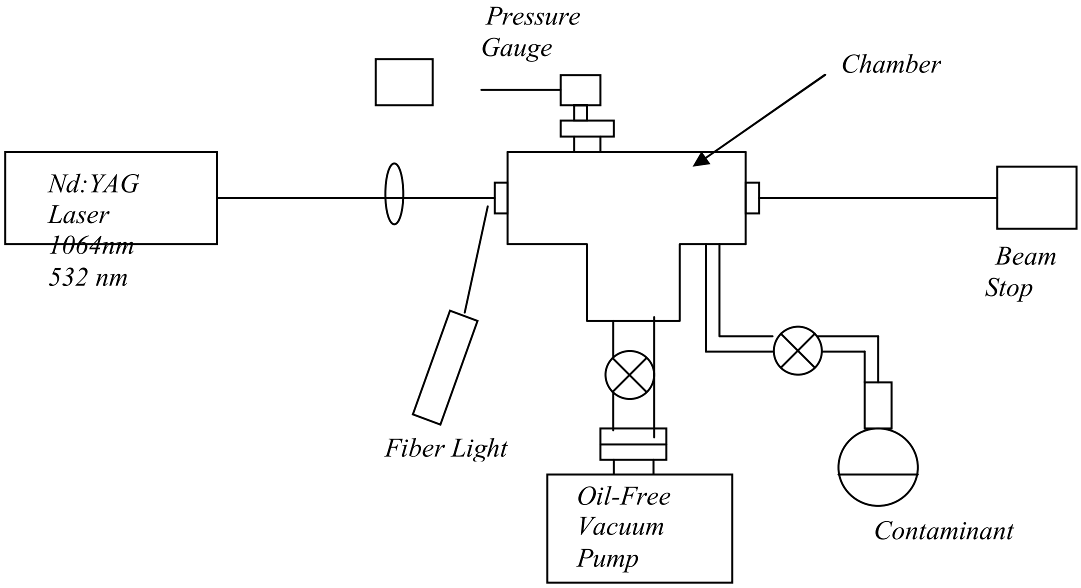


Fig. 1 Block Diagram of Experimental Set-up.

coated windows (CVI, W2-IF-1025-C-1064-0) were made available, and those were also tested under specific conditions. The windows were secured to modified aluminum flanges, and sealed with Viton o-rings.

The 1064 nm laser radiation was generated by a commercial Nd:YAG system (Continuum NY81). The laser was q-switched producing pulse lengths of 10-20 ns, and energies of 170 mJ/pulse, measured with a commercial power meter (Molectron J50 LP2) monitored with an oscilliscope (Tektronix, TDF 340). An amplifier stage was available in this model laser, however it was not used because these laser energies are adequate.

The laser radiation was directed into the chamber using commercial right angle mirrors, and beam is focused with a lens (340 mm f.l.) to a spot size of 5mm on the outer surface of the inlet window. The Guassian profile of the beam leads to a peak laser fluence of approximately 1.5 J/cm^2.

Immediately prior to testing the test sample was cleaned using one of two methods. Initially, the method was a drag wipe using methanol and lens tissue paper. After several tests were run, this was changed such that each sample went through a wet clean process [5] identical to that used on the ICESAT and MLA projects. This process uses Alconox® detergent, and rinses with distilled water and doubly distilled acetone. No marked difference was observed for these experiments between the two methods of cleaning.

After cleaning, the lens was inserted into the test chamber and visually examined using a bright fiber light.

Tests were run on window samples prior to the purposeful addition of any contaminant. Initially, this test was completed in approximately one day, following the practice of Hovis[4], which tallied approximately 400,000 laser shots. Later work verified that the windows could typically endure over 722,000 laser shots. No change to the window could be observed under inspection, so the tests were continued by keeping the same window and adding varying levels of contaminant. Thus, the only variable that changed was the purposeful addition of the contaminant.

III. Results and Discussion

A. Fused Silica

Toluene (HPLC grade, Fischer Scientific) was the first contaminant to be tested. A "positive" result was seen almost immediately, as severe damage to the fused silica substrate was seen to occur quite rapidly (46,000 shots). Typical damage at various levels of severity, is shown in Fig. 2.

To ensure this damage did not result because the substrate was fatigued from the prior 400,000 shots, a new sample was tested immediately adding toluene added to the system. Damage, resembling that labeled moderate, was seen to occur in less than 39,000 counts. This test demonstrated that the toluene was responsible for the rapid onset of damage to the window.

To carry on the research, it was necessary to confirm that the windows routinely held up for 400,000 shots or greater in an uncontaminated environment. Therefore, the chamber was vented, a new window was installed and the system was pumped down, and let to stand overnight. The next morning the system was again pumped down to below 1×10^{-3} torr and the laser was turned on.

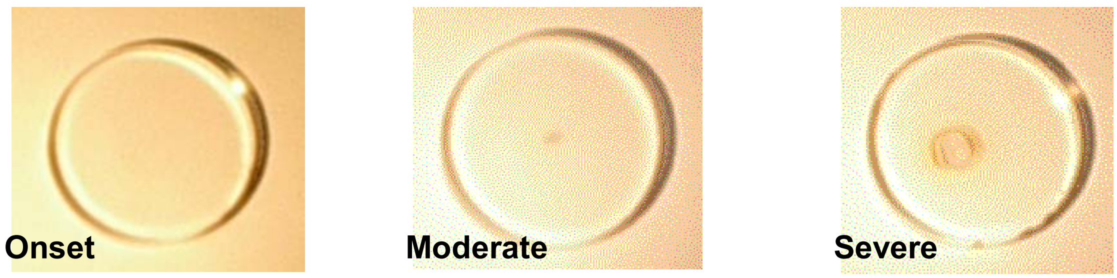
This re-test led to an unexpected but important event. The window for this test began to show damage at just over 106,000 shots. This was much more quickly than observed previously, so another window was installed and tested, and this window damaged in just over 402,000 shots.

These results indicate that toluene remained in the system at trace levels. It is postulated that the toluene adsorbed to the walls, the o-ring seals and other surfaces. Venting and pumping the system at room temperature did not remove the toluene to a level that eliminated the onset of damage. This has profound implications for sealed lasers. It is now confirmed that remediation of compounds used in fabrication is a necessary step in production of sealed high-powered lasers, even those designed to operate in a vacuum.

While this remediation step may vary depending upon the compound, with toluene a straightforward approach was taken. The chamber was vented and pumped out 3-4 times while heated to approximately 40 ^{o}C.

To test whether this was sufficient, another window was tested, and it was shown to endure with no perceptible change for 413,000 shots. At this, the chamber was deemed, “clean” and the tests with toluene ensued.

Fig. 2 Photographs of Damaged Substrates.



The subsequent tests were an effort to monitor the onset of damage as a function of toluene concentration. This onset is the critical element of concern. Once damage begins, it worsens rapidly as the substrate begins to darken and absorb an increasing amount of laser energy; this effect has little if any dependence on the contaminant concentration. On a practical level, once damage starts, the changes in laser properties (power, polarization, divergence, usually deemed essential to the SBL experiment) are dramatic. Moreover, the expected lifetime, that is before the laser ceases operation completely, is likely to be less than a day, depending upon the repetition rate and power of the laser.

These tests were also conducted in the presence of air. The chamber was prepared with toluene as for the vacuum experiments, and then the system was allowed to vent to atmospheric pressure. A negative lens needed to be added to the system to prevent breakdown of the focused laser inside the chamber (the focal point of the lens). It was added immediately in front of the test window so the beam size at the window had only a minimal change.

Under these conditions, no damage was seen to occur with a toluene pressure of 6.8×10^{-2} torr

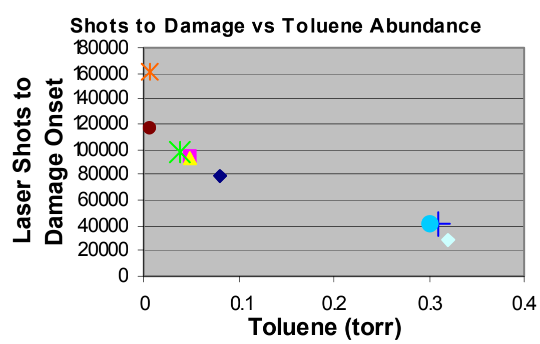
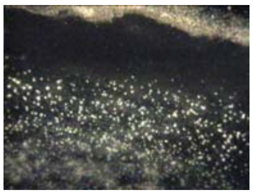


Fig. 3 Chart of onset of damage vs. contaminant concentration.

Moderate Damage
100×

Severe Damage
100×



Moderate Damage
100×

Severe Damage
200×

Fig. 4 Micrographs of laser-induced damage sites.

and over 192,000 laser shots. The test was repeated under vacuum conditions, to determine if the lens had an appreciable effect, and with a toluene pressure of 6.5×10^{-2} torr, the onset of damage was observed in just under 110,000 laser shots.

The onset of damage has been plotted versus the pressure of the toluene, and is presented in Fig. 3. There is an exponential relationship between these two variables.

The damage was inspected using optical microscopy. These images are presented in Fig. 4. The damage starts as a pitting of the surface, which grows in both coverage and depth as the damage worsens. This follows the traditional form of rear surface damage described by Wood [6]. Pitting of this surface is frequently seen, and is explained by an increase in the electric field due to constructive interference from the incident and reflected beams. This interference reaches a maximum at $\lambda/2$ inside the optic ablates material outward.

The system was also tested with acetone instead of toluene. Acetone is commonly used as a solvent in the assembly of spacecraft and to clean laser optics. The previous work cited in Ref. 4, observed light damage using acetone, far less than seen with the toluene.

However, in this work, no damage was seen when using the acetone in the test chamber. The experimental conditions used ranged from 1×10^{-1} torr of acetone exposed to 511,000 laser pulses, to 4.6×10^{-1} torr and over 367,000 laser pulses. These conditions are far more extreme than necessary to see damage for the toluene. While this does not conclusively prove that acetone will not promote laser-induced damage over the lifetime of a SBL instrument, with requirements on the order of a billion shots, it does indicate that this compound is not a high priority of concern. Moreover, its high vapor pressure should make it easy to remediate through traditional means of vacuum bake-outs.

B. MgF_2 Coated Silica

The experiments under vacuum were repeated with the MgF_2 windows. Only six of these windows were available, so the tests were held to a minimum and were expected to yield primarily qualitative results.

A test window, with no contaminant, was seen to last through 426,000 laser shots with no apparent change. When toluene was added to the system at a fairly low concentration, 2.6×10^{-2} torr, severe damage was observed in 56,000 shots. Additional experiments provided similar results.

One sample was tested using acetone in the system. Once again, despite a fairly high pressure of acetone, 1.4×10^{-1} torr, no damage was observed in 344,000 laser shots.

C. Possible Mechanisms

It is worth noting that this damage morphology differs significantly from that seen in Ref. 4. In those works the damage was seen as a build-up of material resembling graphite. Additionally, the damage was seen on the surfaces of optics exposed to toluene, regardless whether it was the front or rear surface. The conclusion from that work was that the laser was initiating a photolytic process that oxidized the C atoms in the toluene to graphite.

In the current work, no build up of any material was observed. Indeed, Electron Spectroscopy for Chemical Analysis (ESCA) was performed to analyze the damaged surfaces, and no carbon species were observed. In this work various mechanisms have been postulated, including that the toluene on the back surface changes the reflectivity at the rear surface. Additional work inspecting an optic with its front surface exposed to the toluene should provide valuable information regarding this possible mechanism.

Another mechanism postulated is that the toluene is undergoing photolysis in the gas phase, and that these species react with the surface. Abundant work[7] has been performed showing the photodissociation and photoionization of toluene and toluene clusters. The variety of species formed includes a number of reactive chemical radicals and ions. It is conceivable that these species are reactive enough to etch the window surface, particularly in the presence of laser light. The confirmation of such a mechanism would be highly challenging.

This MgF_2 research provides a vital element in discerning the mechanism. One potential mechanism was that the toluene had a unique chemical interaction with the SiO_2 surface. This interaction would then lead to greater absorption of the laser light and damage due to heating, or it would lead to chemically reactive species that etched the silica.

However, it is highly unlikely that MgF_2, with different elements and a different crystalline structure, would offer that same interaction with toluene. Yet, damage occurs with the same morphology as in the silica. This points to the phenomenon being either one involving gas-phase toluene or the change in the index of refraction at the surface

Finally, it is worth mentioning that a few of the tests showed no damage in the presence of toluene, despite being exposed to more laser pulses than was seen to initiate damage in other samples. While these results can be removed from the mathematical data analysis using Student's t-test, this too may provide a clue as to the mechanism.

Wood reports in Ref. 6 that the surface preparation has an enormous effect on the laser-induced damage threshold. It could be that a few optics had much better surfaces (ie fewer scratches or defects) than the majority, so their thresholds were higher. This is probably most consistent with the change in index of refraction mechanism. If the damage was due to the gas-phase species one would still expect damage, as it seems unlikely that a polished surface would become so chemically inert.

IV. Conclusions

A working test facility has been constructed to test for laser-induced damage to optics promoted by trace, gas-phase contaminants in sealed lasers. Test optics included fused silica windows, and MgF_2 coated windows. Both produced consistent results in observing damage.

Toluene was found to promote this damage to a high degree, and should be treated with concern for such systems. Toluene also serves as a representative aromatic compound, and all of these compounds should be scrutinized. Acetone however, showed no indication of promoting damage in the limited number of sample runs performed.

No clear mechanism for the promotion of damage has been confirmed, although this work has offered some direction in this area. Looking exclusively at a surface/contaminant (photo)chemical interaction seems to be unlikely.

As demonstrated in the literature, the presence of air (likely molecular oxygen) seems to eliminate this effect. However, in contrast to this work, the morphology of the damage is dramatically different.

Finally, the test plan can and is being used to test non-volatile compounds, such as adhesives, that are known to have been used on current SBL instruments. This should provide insight into concerns for these missions, and help to address design concerns for future SBL missions.

V. References

[1] http://icesat.gsfc.nasa.gov/

[2] http://ltpwww.gsfc.nasa.gov/tharsis/mola.html
http://ltpwww.gsfc.nasa.gov/ltp/mla.html

[3] J. L. LaCour, et al, "Laser Induced Breakdown Spectrocopy under Martian Conditions." Poster 1582, Thirty-fourth Lunar and Planetary Science Conference, Houston, TX, 2003
K. Knight, et al, "Development of a Prototype Instrument for Stand-off Elemental Analysis for use on a Mars Rover." Found at http://marsprogram.jpl.nasa.gov/mgs/sci/fifthconf99/6064.pdf

[4] F. E. Hovis, B. Shepherd, C. Radcliffe and H. Maliborski, "Mechanisms of Contamination Induced Optical Damage in Lasers." Proceedings of the SPIE, vol. 2428, pp. 72-83, 1995.
F. E. Hovis, B. A. Shepherd, C. T. Radcliffe and H. A. Maliborski, "Contamination damage in pulsed 1 μm lasers." Proceedings of SPIE, vol. 2714, pp. 707-716, 1996.
F. E. Hovis, B.A. Shepherd, C.T. Radcliffe, A.L. Bailey and W.T. Boswell, "Optical damage at the part per million level: The role of trace contamination in laser induced optical damage." Proceedings of SPIE, vol. 2114, pp 1-8, 1994.

[5] GSFC Work Instruction 551-WI-8072.1.7.

[6] R. M. Wood, *Laser-Induced Damage of Optical Materials*. Bristol: Institute of Physics Publishing, 2003.

[7] D. W. Squire and R. B. Bernstein, "Multiphoton Ionization Mass Spectrometric Study of Toluene Clusters in a Pulsed Nozzle Beam Time-of-Flight Apparatus." *J Chem Phys* vol. 88, pp 4944-4952, 1984.

L. D. Brouwer, W. Muller-Markgraf and J. Troe, "Thermal Decomposition of Toluene: A Comparison of Thermal and Laser Photochemical Activation Experiments." *J Chem Phys* vol. 92, pp. 4905-4914, 1988.

R. Frochtenicht, "The photodissociation of toluene studied by forward photofragment translational spectroscopy." *J Chem Phys* vol. 102, pp. 4850-4859, 1995.